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<b>UTILITY PATENT APPLICATION TRANSMITTAL</b> (Only for new nonprovisional applications under 37 C.F.R. § 1.53(b))	Attorney Docket No. <u>97-2166</u>
	First Inventor or Application Identifier <u>Chu</u>
	Title <u>Ceramic Particles Formed In-Situ</u>
	Express Mail Label No. <u>EJ244060992 US</u>

<b>APPLICATION ELEMENTS</b> See MPEP chapter 600 concerning utility patent application contents.	<b>ADDRESS TO:</b> Assistant Commissioner for Patents Box Patent Application Washington, DC 20231
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1. <input checked="" type="checkbox"/> * Fee Transmittal Form (e.g., PTO/SB/17) (Submit an original and a duplicate for fee processing) 2. <input checked="" type="checkbox"/> Specification [Total Pages <u>24</u> ] - Descriptive title of the invention - Cross References to Related Applications - Statement Regarding Fed sponsored R & D - Reference to Microfiche Appendix - Background of the invention - Brief Summary of the invention - Brief Description of the Drawings (if filed) - Detailed Description - Claim(s) - Abstract of the Disclosure 3. <input checked="" type="checkbox"/> Drawing(s) (35 U.S.C. 113) [Total Sheets <u>4</u> ] 4. Oath or Declaration [Total Pages <u>2</u> ] a. <input type="checkbox"/> Newly executed (original or copy) b. <input checked="" type="checkbox"/> Copy from a prior application (37 C.F.R. § 1.63(d)) (for continuation/divisional with Box 17 completed) [Note Box 5 below] i. <input type="checkbox"/> DELETION OF INVENTOR(S) Signed statement attached deleting inventor(s) named in the prior application, see 37 C.F.R. §§ 1.63(d)(2) and 1.33(b). 5. <input checked="" type="checkbox"/> Incorporation By Reference (useable if Box 4b is checked) The entire disclosure of the prior application, from which a copy of the oath or declaration is supplied under Box 4b, is considered to be part of the disclosure of the accompanying application and is hereby incorporated by reference therein.	6. <input type="checkbox"/> Microfiche Computer Program (Appendix) 7. Nucleotide and/or Amino Acid Sequence Submission (if applicable, all necessary) a. <input type="checkbox"/> Computer Readable Copy b. <input type="checkbox"/> Paper Copy (identical to computer copy) c. <input type="checkbox"/> Statement verifying identity of above copies
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ACCOMPANYING APPLICATION PARTS	
8. <input type="checkbox"/> Assignment Papers (cover sheet & document(s))	
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**\* NOTE FOR ITEMS 1 & 14: IN ORDER TO BE ENTITLED TO PAY SMALL ENTITY FEES, A SMALL ENTITY STATEMENT IS REQUIRED (37 C.F.R. § 1.27), EXCEPT IF ONE FILED IN A PRIOR APPLICATION IS RELIED UPON (37 C.F.R. § 1.28).**

17. If a CONTINUING APPLICATION, check appropriate box, and supply the requisite information below and in a preliminary amendment:

☐ Continuation ☒ Divisional ☐ Continuation-in-part (CIP) of prior application No: 08, 978, 221

Prior application information: Examiner T. McGuthry Banks Group / Art Unit: 1742

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of )  
Men G. Chu et al ) Examiner T. McGuthry Banks  
Serial No. not yet received ) Group Art Unit 1742  
Filed concurrently herewith )  
For Method of Forming Ceramic Particles ) Alcoa Docket 97-2166  
In-Situ in Metal )

**Preliminary Amendment**

I hereby certify that this correspondence is being deposited  
with the United States Postal Service as first class mail in  
an envelope addressed to: Assistant Commissioner for  
Patents, Washington, D.C. 20231 on  
July 9, 1999

  
David W. Pearce-Smith, Reg. No. 31285  
Date of Signature: July 9, 1999

Assistant Commissioner for Patents  
Box Patent Application  
Washington, D.C. 20231

Sir:

Please amend the above-identified application as follows:

Please delete the original title and substitute therefor --Metal Product

Containing Ceramic Dispersoids Formed In-Situ--.

In the specification:

Page 1, before the first line, insert the following paragraph:

-- Cross Reference to Related Applications

This application is a division of application Serial No. 08/978,221, filed on November 25, 1997. --

In the claims:

Please cancel claims 1-17.

Amend the claims as follows:

Claim 18, line 3, delete "ins-situ" and substitute --in-situ--.

Remarks

Claims 1-17 as are currently being prosecuted in the parent application have been cancelled herein. Remaining method claims 18-21 do not contain new matter.

Respectfully submitted,

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## CERAMIC PARTICLES FORMED IN-SITU IN METAL

### Technical Field Of The Invention

The present invention relates to fine ceramic particles formed in-situ in metal and in alloys by the liquid-state process, and to products containing the fine ceramic particles formed in-situ in metal and in alloys by the liquid-state process. In one aspect, the present invention relates to a process for producing a material containing uniformly dispersed, finely sized ceramic phase particles, e.g., such as titanium carbide particles, formed in-situ in metals and in alloys by the liquid-state process.

### Background of the Invention

The aluminum and aerospace industries have long sought a method to control recrystallization of aluminum alloys during deformation operations to permit the design of aluminum airframes with improved structural properties.

The metals industry today conventionally uses dispersoids, i.e., fine particles dispersed in the metal alloy, to control recrystallization and to increase dispersion strengthening at elevated temperatures. Such dispersoids of fine particles dispersed in the metal alloy usually are formed by solid state precipitation.

Recent developments in this area suggest that to improve formability and high temperature strength of aluminum alloys, it is necessary to increase the number densities and to reduce the size of the fine particle size dispersoids.

Certain emerging technologies are capable of producing fine particulates of different types with somewhat improved interfacial characteristics. Among the several

ways of producing these materials, the technologies where the particles are introduced or formed in the molten aluminum prior to its solidification are attractive, primarily because of the potential for commercially economic processes on a large scale.

A variety of processing routes classified generally as in-situ ceramic phase formation processes in metal have been developed recently. According to the state of the reactants in the process, such a ceramic phase formation process in metal generally is classified into one of several categories:

- (1) liquid metal - gas reaction,
- (2) liquid metal - liquid metal reaction, or
- (3) liquid - solid reaction.

In the case of carbon particles or carbon blocks in the context of liquid metal - liquid metal reactions or liquid - solid reactions, it is known that such carbon particles or carbon blocks are difficult to introduce directly into a melt in metal because of non-wetting of the carbon by the molten metal or alloy.

### **Introduction to the Invention**

Recent developments in liquid metal - gas reaction processes have produced fine TiC particulates in a molten aluminum alloy. In this approach, a carbonaceous gas is introduced into an aluminum melt containing titanium to form TiC particulates, and the carbide volume fraction is determined by the initial titanium content. When the melt containing the carbides is cast and subsequently extruded for microstructure and property evaluation, the as-cast microstructure of the in-situ processed composites reveals a

relatively uniform distribution of TiC particles with an average size of a few microns. No preferential particle segregation is observed in the dendritic cell boundaries generally.

U.S. Patent 4,808,372, issued to Koczak et al., discloses an in-situ process for producing a composite containing refractory material. A molten composition,  
5 comprising a matrix liquid, and at least one refractory carbide-forming component are provided, and a gas is introduced into the molten composition. Methane is bubbled through a molten composition of powdered aluminum and powdered titanium to produce a carbide having an average particle size in the fine mode of about 3 to 7 microns and in the coarse mode of about 35 microns.

Although conventional ceramic phase formation processes in metal offer  
10 some possibilities for the production of a wide range of reinforcement particle types and improved compatibility between the reinforcement and the matrix, the in-situ formed ceramic particles in metal are too large, e.g., on the order of several microns, and tend to form clusters. In-situ formed ceramic particles having these sizes, i.e., of several  
15 microns, are candidates for use as reinforcement in a composite, but are not suitable for use as dispersoids for recrystallation control, for dispersion strengthening, or for use as a component for structure refinement.

Accordingly, a novel ceramic dispersoid in metal product and process for making such a novel ceramic dispersoid in metal product are needed for providing  
20 uniformly dispersed, finely sized ceramic phase particles dispersed in-situ in a metal matrix.

U.S. Patents 4,842,821 and 4,748,001, issued to Banerji et al., disclose a method for producing a metal melt containing dispersed particles of titanium carbide.

Carbon particles are reacted with titanium in the metal to obtain titanium carbide. The patent discloses that salts preferably are entirely absent from the melt (U.S. Patent

5 4,842,821, col. 3, lines 26-28, and U.S. Patent 4,748,001, col. 3, lines 40-42).

U.S. Patent 5,405,427, issued to Eckert, discloses a flux composition for purifying molten aluminum to remove or capture inclusions in the melt and carry such inclusions to the surface (col. 4, line 13 et seq.). The flux composition contains sodium chloride, potassium chloride, and a minor amount of magnesium chloride and carbon particles.

U.S. Patent 5,401,338, issued to Lin, discloses a process for making metal matrix composites wherein fine particles (0.05 microns) of alumina, silicon nitride, silicon carbide, titanium carbide, zirconium oxide, boron carbide, or tantalum carbide are added into a metal alloy matrix (col. 2, lines 64-68).

U.S. Patent 5,041,263, issued to Sigworth, discloses a process for providing a grain refiner for an aluminum master alloy that contains carbon or other third elements and acts as an effective refiner in solution in the matrix, rather than being present as massive hard particles.

Uniformly high number densities of finely sized dispersoids increase the recrystallization temperature, inhibit grain growth in hot working, and improve elevated temperature strength. Further, fine particles of dispersoids are effective nuclei for grain

refining.

It is against this need in the background technology that the present invention was made.

Accordingly, it is an object of this invention to provide aluminum alloys  
5 having high number densities of fine ceramic particles to act as dispersoids.

Accordingly, it is an object of the present invention to provide a method for increasing the number densities of dispersoids in the liquid state and which then remain stable and dispersed in the solid state in metal alloys.

It is an object of the present invention to produce finely sized ceramic phase  
10 particles.

It is a further object of the present invention to produce uniformity in the dispersion of finely sized ceramic phase particles in metal and in alloys.

It is yet another object of the present invention to produce uniformly distributed, finely sized ceramic phase particles dispersed in-situ in a metal matrix.

It is another object of the present invention to produce uniformly  
15 distributed, finely sized ceramic phase particles dispersed in-situ in a metal alloy in a process providing reaction times shorter than conventional approaches.

It is another object of the present invention to produce uniformly distributed, finely sized ceramic phase particles dispersed in-situ in a metal alloy for  
20 recrystallization control, dispersion strengthening, or grain refining.

These and other objects of the present invention will become apparent from



the detailed description which follows.

### **Summary of the Invention**

The present invention provides a novel method for producing a ceramic phase particle dispersoid in metal and a novel product composed thereof. The method includes (a) providing a molten composition consisting essentially of molten aluminum alloy and molten metal selected from the group consisting of Zr, V and combinations thereof; (b) providing a chloride salt containing fine carbon particles; and (c) reacting the chloride salt containing fine carbon particles in the molten aluminum metal liquid with the molten metal liquid to form a uniform distribution of finely sized carbide particles formed and dispersed in-situ in an aluminum alloy matrix.

The step of reacting includes vigorously stirring the mixture containing salt, metal alloy, and carbon to form a reaction mixture at a temperature above the liquidus of alloy to form a uniform distribution of particles sized less than about 2.5 microns uniformly dispersed in-situ in the metal matrix.

Another embodiment of the invention is a ceramic dispersoid in metal product comprising: (a) a matrix metal; and (b) a uniform distribution of formed and dispersed in-situ in the metal matrix. The finely sized ceramic particles are formed by the process of: (i) providing a molten composition comprising a matrix liquid of aluminum or aluminum alloy metal and at least one carbide-forming element selected from the group consisting of Ti, Sc, Hf, Nb, Zr, Mo, and V; (ii) providing a chloride salt containing carbon particles, wherein the salt comprises NaCl and KCl in a weight/weight

ratio within the range of about 0.8-1.2 and of  $MgCl_2$  and  $CaCl_2$  in amounts comprising up to about 5-10% by weight of the salt mixture; and (iii) reacting the chloride salt containing carbon particles in the molten aluminum alloy by vigorously stirring the aluminum alloy and the chloride salt containing carbon particles to form a mixture of the molten metal liquid in contact with a portion of the carbon particles at an elevated temperature above the liquidus of the aluminum alloy to form a unagglomerated distribution of finely sized ceramic phase particles having an average particle diameter of less than about 0.3 microns formed and dispersed in-situ in an aluminum metal matrix.

### **Brief Description of the Drawings**

Figure 1 shows a photomicrograph of a ceramic dispersoid in metal as produced by conventional processes available in the prior art.

Figure 2 shows a photomicrograph of a ceramic dispersoid in metal as produced and provided by the present invention.

Figure 3 is a DTA showing the temperature of formation of  $TiC$ .

Figure 4 is a DTA showing the temperature of formation of  $ZrC$ .

Figure 5 is a DTA showing the temperature of formation of  $VC$ .

### **Detailed Description**

The terms "fine" and "finely sized" as it refers to intermetallic particles is a particle being less than about 2.5 microns.

The present invention provides a novel liquid-state dispersoid-forming process, novel ceramic particle dispersoids formed in-situ in metal by the liquid-state

process, and novel products containing the ceramic particle dispersoids formed in-situ in metal by the liquid-state process. In one aspect, the present invention provides a novel product and process for producing a material containing uniformly dispersed, finely sized ceramic phase particles, e.g., such as titanium carbide particles, formed in-situ in metal by the liquid-state dispersoid-forming process.

In one aspect, the novel ceramic dispersoid in metal product and process for producing such a ceramic dispersoid in metal include uniformly dispersed and finely sized carbide particles of the present invention formed in-situ in metal. In this one aspect, the present invention incorporates a novel mixing process involving the following two components:

(1) molten metal in combination with at least one of the carbide-forming elements including Ti, Sc, V, Hf, Nb, Zr, Mo, and Al (when the molten metal matrix is not aluminum); and

(2) salt containing fine carbon particles or dissolved carbon or a combination of fine carbon particles and dissolved carbon.

The present invention includes controlling and selecting the liquidus temperature of the salt to a value lower than that of the molten metal. The present invention further includes controlling and selecting the salt for the purpose of wetting the carbon particles.

The present invention includes a specific mixing process, wherein a first component of molten metal containing carbide-forming elements is provided. A second

component, either in the solid or molten state, initially is added to the first component of molten metal containing carbide-forming elements. When both first and second components are in the liquid state, the melt is vigorously stirred mechanically or electromagnetically over a period of time. During the stirring, the salt is finely dispersed, and the process of the present invention provides for the carbon to react with the carbide-forming element substantially instantaneously to form fine carbide particles. After reaction, the salt is decanted or removed.

The melt is then alloyed with any desirable alloying elements.

The alloy melt containing fine carbide particles is then cast into a mold, or cast to form ingot (rectangular or round), slab, sheet, or strip. The alloy melt can be spray formed to form bulk product.

The molten salt used for the process of the present invention enhances the reaction of carbon and the carbide-forming component in the alloy. The molten salt provides that the alloy is cleaned of any oxide or dross and, hence, a fresh surface is available for reaction. Carbon has some small but finite solubility in the molten salt. As reaction proceeds, the salt is depleted with respect to carbon. Hence, more carbon is dissolved, and the dissolved carbon reacts with the carbide forming element in the alloy to produce the fine particulates of carbides of the present invention. In accordance with the present invention, the carbon does not necessarily have to be dissolved in the molten salt for reaction to occur. Fine particulates of carbon also can take part in the reaction. Moreover, all of the carbon to be reacted need not be suspended in the salt at one time.

Only a portion of the carbon need be in reactive contact, and when that carbon reacts, more carbon is brought into reaction contact by the vigorous stirring of the present invention.

The specific choice of salt composition in accordance with the present invention involves a molten salt containing elements which will not contaminate the metal by way of reacting with aluminum metal or aluminum alloying elements. The specific choice of salt composition in accordance with the present invention involves a salt which is thermodynamically stable and compatible with the metal. The present invention selects from the halide salts of alkali and alkaline earth metals. The halides of Na, K, Ca, Mg, and Li are preferred. Eutectic melts of binary, ternary, or quaternary salts with or without other additives may be used. The salt also preferably has a melting point below about 900°C and, more preferably, below about 600°C. The eutectic melts of NaCl-KCl with small additions of  $MgCl_2$  and  $CaCl_2$  are particularly preferred. The NaCl and KCl weight/weight ratio should be about 1.0, preferably within 0.8-1.2. The additives of  $MgCl_2$  and  $CaCl_2$  preferably make up about 5-10% by weight of the salt mixture in accordance with the present invention.

In one aspect, the present invention employs a salt containing the following constituents and approximate percentages by weight, most preferably, NaCl: 48%, KCl: 48%,  $MgCl_2$ : 2.2%, and  $CaCl_2$ : 1.8%. This salt has a eutectic of about 600°-645°C, most preferably, of about 645°C.

The salt system of the present invention preferably has a eutectic capable of

dissolving at a temperature below the liquidus of the metal matrix, e.g., in one aspect below the liquidus of aluminum is workable.

In addition, salts of  $\text{MgCl}_2$ -KCl,  $\text{MgCl}_2$ -NaCl, KCl- $\text{CaCl}_2$ -NaCl also can be used in the system in accordance with the present invention. Salts having the eutectic composition and with the specified melting points will be preferred.

In addition, molten salts containing fluorides of Na, Ca, K, Mg, and Li can be used in the system in accordance with the present invention. When these fluoride salts are used, special care should be taken to provide that no excessive fluorides are evolved during the processing.

Although the process is described for carbides only, it can be extended to:

1. borides by incorporating boron in place of carbon,
2. nitrides, by incorporating nitrogen compounds such as AlN in place of carbon and
3. similar such refractory material compounds having relatively high melting temperatures and hardness, and relatively low chemical reactivity in comparison to non-refractory materials.

The present invention provides for the formation of fine carbide particles in metal. The particles produced in situ in metal in accordance with the present invention are well-dispersed in the metal. The process in accordance with the present invention includes mixing a molten metal of a carbide-forming element with a low liquidus temperature salt containing fine carbon particles or dissolved carbon. Both components

are brought to reactive contact in the liquid state and thoroughly mixed. After reaction of carbon with carbide-forming element, the salt is decanted or removed. The melt which contains uniformly distributed, finely sized, unagglomerated carbide particles is cast into a mold or cast to form ingot and the like.

5 Referring now to Figure 1, a section of casting is shown in microstructure by actual photomicrograph. A ceramic dispersoid in metal as produced by conventional processes available in the prior art is shown. Large size particles in uneven dispersion are apparent.

10 Referring now to Figure 2, a section is shown of the uniformly dispersed finely sized titanium carbide particles formed in situ in aluminum in accordance with the present invention. The particles are observed in microstructure to be finely sized with an average particle diameter less than about 0.3 microns and can be seen to be uniformly dispersed throughout the metal.

15 It has been found empirically that the present invention produces uniformly dispersed, finely sized ceramic phase particles formed and dispersed in-situ in a metal matrix. It has been found further that the present invention produces uniformly dispersed, finely sized ceramic phase particles formed and dispersed in-situ in a metal matrix in a process requiring reaction times shorter than existing conventional approaches, e.g., on the order of less than about one hour. The uniformly dispersed, finely sized ceramic  
20 phase particles dispersed in-situ in a metal matrix are suitable not only for application of reinforcement in a composite, but also for recrystallization control, dispersion

strengthening, or grain refining.

### Example 1

A first component melt of 1.5 Kg of aluminum - 2 % titanium (1016 grams Al, 484 grams Ti) provided by Aluminum Company of America, Alcoa Technical Center, Alcoa Center, Pennsylvania, was prepared and heated to about 983°C. A second component mixture (922 grams total) of carbon particles and a salt (700 grams) containing about 48% NaCl, 48% KCl, 2.2% MgCl<sub>2</sub>, and 1.8% CaCl<sub>2</sub> by weight was prepared and heated to about 200 F overnight. The preheated first and second components were added together in a crucible and heated to a temperature of about 983°C.

A mechanical stirring was applied by graphite propeller inserted into the crucible. A lid was placed to cover the crucible during reaction and to permit insertion of the graphite propeller and a thermocouple. After vigorous stirring and reaction for 15 minutes, the salt was skimmed, and the melt was cast into 1.5 inch diameter graphite molds. After cooling, the casting was cut for characterization.

The structure of the casting is shown in Figure 2. As shown, the fine TiC particles are as small as submicrons in size and uniformly dispersed in the matrix.

The micro-composite particles of TiC in accordance with the present invention increase the ambient temperature strength and the elastic modulus of the aluminum base alloy.



### Example 2

A mixture of 2.6 grams of Al-6% Ti powder and 0.32 grams graphite powder. 0.72 grams was formed and then placed in an  $\text{Al}_2\text{O}_3$  crucible of a DTA unit (Differential Thermal Analyzer). The crucible and sample was then heated to a temperature from room temperature  $1300^\circ\text{C}$  at a rate of  $50^\circ\text{C}/\text{min}$  under an argon atmosphere.

The results of the DTA is shown in Figure 3. A peak shown at approximately  $1275^\circ\text{C}$  (as indicated by an arrow in Figure 3) is an exothermic reaction, indicating the formation of TiC particles.

### Example 3

The procedure of Example 2 was repeated except that 2.6 grams of Al-5.7% Zr powder was mixed with 0.32 grams graphite powder. 0.71 grams. As before the mixture placed into an  $\text{Al}_2\text{O}_3$  crucible of a DTA unit and heated to a temperature  $1300^\circ\text{C}$  at a rate of  $50^\circ\text{C}/\text{min}$  under an argon atmosphere.

The results of the DTA is shown in Figure 4. A peak shown at approximately  $1275^\circ\text{C}$  (as indicated by an arrow in Figure 4) is an exothermic reaction, indicating the formation of ZrC particles. These ZrC particles act as a dispersoid in aluminum alloys.

### Example 4

The procedure of Example 2 was repeated except that 2.6 grams of Al-5.1% V powder was mixed with 0.32 grams graphite powder. 0.71 grams of this mixture was

then placed in an  $\text{Al}_2\text{O}_3$  crucible of a DTA unit. The sample was heated to a temperature 1300°C at a rate of 50°C/min under an argon atmosphere.

The results of the DTA is shown in Figure 5. A peak shown at approximately 1270°C (as indicated by an arrow in Figure 5) is an exothermic reaction, indicating the formation of VC particles. These VC particles act as a dispersoid in aluminum alloys.

While the invention has been described in conjunction with several embodiments, it is to be understood that many alternatives, modifications, and variations will be apparent to those skilled in the art in light of the foregoing description.

Accordingly, this invention is intended to embrace all such alternatives, modifications, and variations which fall within the spirit and scope of the appended claims.

What is claimed is:

1. A method of forming finely sized carbide phase particles formed in situ in a molten metal or molten metal alloy, comprising:

(a) providing a molten composition consisting essentially of molten aluminum alloy and molten metal selected from the group consisting of Zr, V and combinations thereof;

(b) providing a chloride salt containing fine carbon particles; and

(c) reacting said chloride salt containing fine carbon particles in said molten aluminum metal liquid with said molten metal to form a uniform distribution of finely sized carbide particles formed and dispersed in-situ in an aluminum alloy matrix.

2. The method as set forth in claim 1 wherein said step of reacting said chloride salt containing carbon particles in said molten aluminum comprises vigorously stirring said molten composition and said chloride salt containing carbon particles to form a mixture of said molten metal in contact with a portion of said carbon particles at an elevated temperature for sufficient residence time to form a uniform distribution of finely sized (ceramic phase) metal carbide particles formed and dispersed in-situ in a metal matrix.

3. The method as set forth in claim 2 wherein said finely sized metal carbide particles comprise titanium carbide particles having an average particle diameter of less than about 0.3 microns formed in situ in metal.

4. The method as set forth in claim 2 wherein said finely sized metal carbide particles are selected from the group consisting of ZrC, VC and combinations thereof.

5. The method as set forth in claim 2 further comprising:

(d) controlling and selecting said salt to have a liquidus temperature lower than that of said molten aluminum metal liquid.

6. The method as set forth in claim 5 wherein said step of controlling and selecting said salt further comprises selecting said salt for the purpose of wetting said carbon particles.

7. The method as set forth in claim 6 wherein said residence time is less than one hour.

8. The method as set forth in claim 6 wherein said salt comprises chloride salts of alkali and alkaline earth metals.

9. The method as set forth in claim 8 wherein said salt comprises a eutectic melt of NaCl-KCl with minor amounts of  $\text{MgCl}_2$  and  $\text{CaCl}_2$ .

10. The method as set forth in claim 9 wherein said salt has a melting point below about  $600^\circ\text{C}$ .

11. The method as set forth in claim 10 wherein said salt has a NaCl and KCl weight/weight ratio within the range of about 0.8-1.2, and the additives of  $\text{MgCl}_2$  and  $\text{CaCl}_2$  comprise up about 5-10% by weight of the salt mixture.

12. The method as set forth in claim 11 wherein said salt has a eutectic of about  $600\text{-}700^\circ\text{C}$ .

13. The method as set forth in claim 11 wherein said salt contains about 48% NaCl, 48% KCl, 2.2%  $\text{MgCl}_2$ , and 1.8%  $\text{CaCl}_2$  by weight.

14. A method of forming finely sized carbide phase particles formed in situ in a molten aluminum metal or aluminum metal alloy comprising:

(a) providing a molten composition consisting essentially of molten aluminum alloy and molten metal selected from the group consisting of Zr, V and combinations thereof;

(b) providing a chloride salt containing carbon particles, wherein said salt comprises NaCl and KCl in a weight/weight ratio within the range of about 0.8-1.2 and of  $MgCl_2$  and  $CaCl_2$  in amounts comprising up to about 5-10% by weight of the salt mixture; and

(c) reacting said chloride salt containing carbon particles in said molten aluminum alloy by vigorously stirring said aluminum alloy and said chloride salt containing carbon particles to form a mixture of said molten metal liquid in contact with a portion of said carbon particles at an elevated temperature above the liquidus of the aluminum alloy to form a unagglomerated distribution of finely sized ceramic phase particles having an average particle diameter of less than about 0.3 microns formed and dispersed in-situ in an aluminum metal matrix.

15. The method as set forth in claim 14 wherein said finely sized metal carbide particles are selected from the group consisting of  $ZrC$ ,  $VC$  and combinations thereof.

16. A method of forming finely sized carbide phase particles formed in situ in a molten aluminum metal or aluminum metal alloy comprising:

(a) providing a molten composition comprising a matrix liquid of aluminum or aluminum alloy metal and at least one carbide-forming element selected from the group consisting of Ti, Sc, Hf, Nb, Zr, Mo, and V;

(b) providing a chloride salt containing carbon particles, wherein said salt comprises NaCl and KCl in a weight/weight ratio within the range of about 0.8-1.2 and of  $MgCl_2$  and  $CaCl_2$  in amounts comprising up to about 5-10% by weight of the salt mixture; and

(c) reacting said chloride salt containing carbon particles in said molten aluminum alloy by vigorously stirring said aluminum alloy and said chloride salt containing carbon particles to form a mixture of said molten metal liquid in contact with a portion of said carbon particles at an elevated temperature above the liquidus of the aluminum alloy to form a unagglomerated distribution of finely sized ceramic phase particles having an average particle diameter of less than about 0.3 microns formed and dispersed in-situ in an aluminum metal matrix.

17. The method as set forth in claim 16 wherein said finely sized metal carbide particles are selected from the group consisting of ZrC, VC and combinations thereof.

18. A ceramic dispersoid in metal product, comprising:

(a) a matrix metal and

(b) a uniform distribution of formed and dispersed ins-situ in said metal

matrix.



19. The ceramic dispersoid in metal product of claim 18 wherein said finely sized ceramic particles are formed by the process of:

(a) providing a molten composition comprising a matrix liquid of aluminum or aluminum alloy metal and at least one carbide-forming element selected from the group consisting of Ti, Sc, Hf, Nb, Zr, Mo, and V;

(b) providing a chloride salt containing carbon particles, wherein said salt comprises NaCl and KCl in a weight/weight ratio within the range of about 0.8-1.2 and of  $MgCl_2$  and  $CaCl_2$  in amounts comprising up to about 5-10% by weight of the salt mixture; and

(c) reacting said chloride salt containing carbon particles in said molten aluminum alloy by vigorously stirring said aluminum alloy and said chloride salt containing carbon particles to form a mixture of said molten metal liquid in contact with a portion of said carbon particles at an elevated temperature above the liquidus of the aluminum alloy to form a unagglomerated distribution of finely sized ceramic phase particles having an average particle diameter of less than about 0.3 microns formed and dispersed in-situ in an aluminum metal matrix.

20. The ceramic dispersoid in metal product of claim 18 wherein said finely sized metal carbide particles are selected from the group consisting of ZrC, VC and combinations thereof.

21. The ceramic dispersoid in metal product of claim 18 wherein said matrix metal is aluminum or an aluminum alloy.

### Abstract of the Disclosure

A novel method for producing a ceramic phase particle dispersoid in metal and a novel product composed thereof. The method includes (a) providing a molten composition consisting essentially of molten aluminum alloy containing molten metal selected from the group consisting of Zr, V and combinations thereof; (b) providing a chloride salt containing fine carbon particles; and (c) reacting the chloride salt containing fine carbon particles in the molten aluminum metal liquid with the molten metal liquid to form a uniform distribution of finely sized carbide particles formed and dispersed in-situ in an aluminum alloy matrix.

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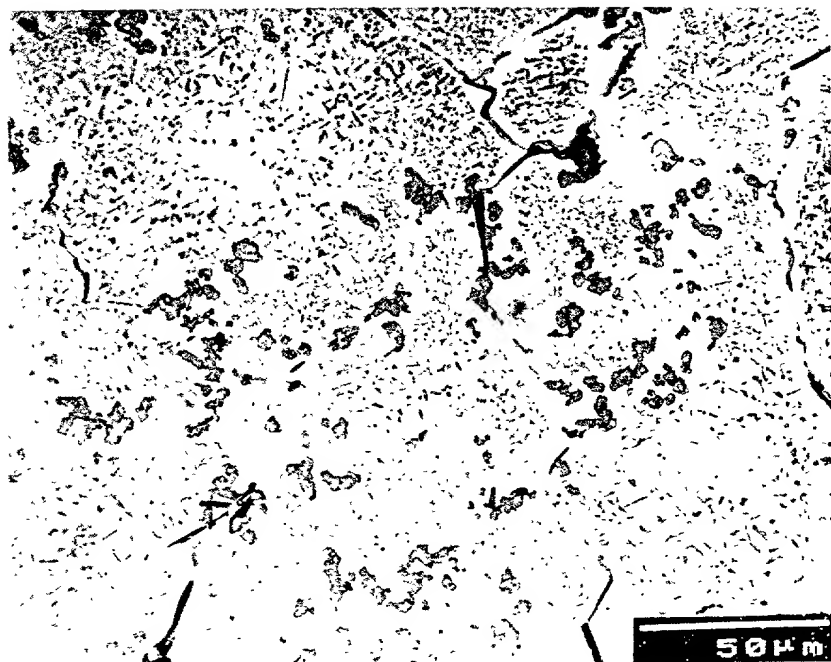


FIG. 1

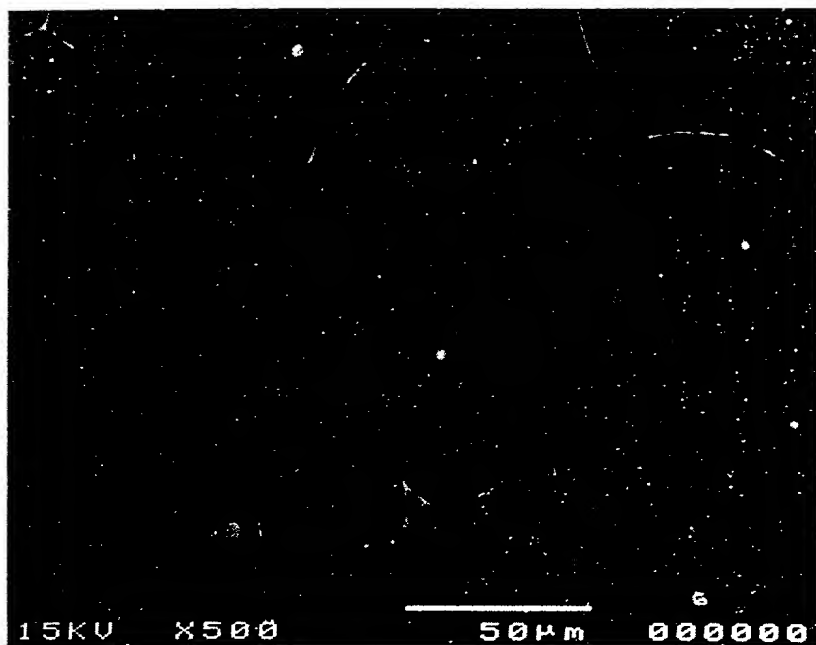


FIG. 2

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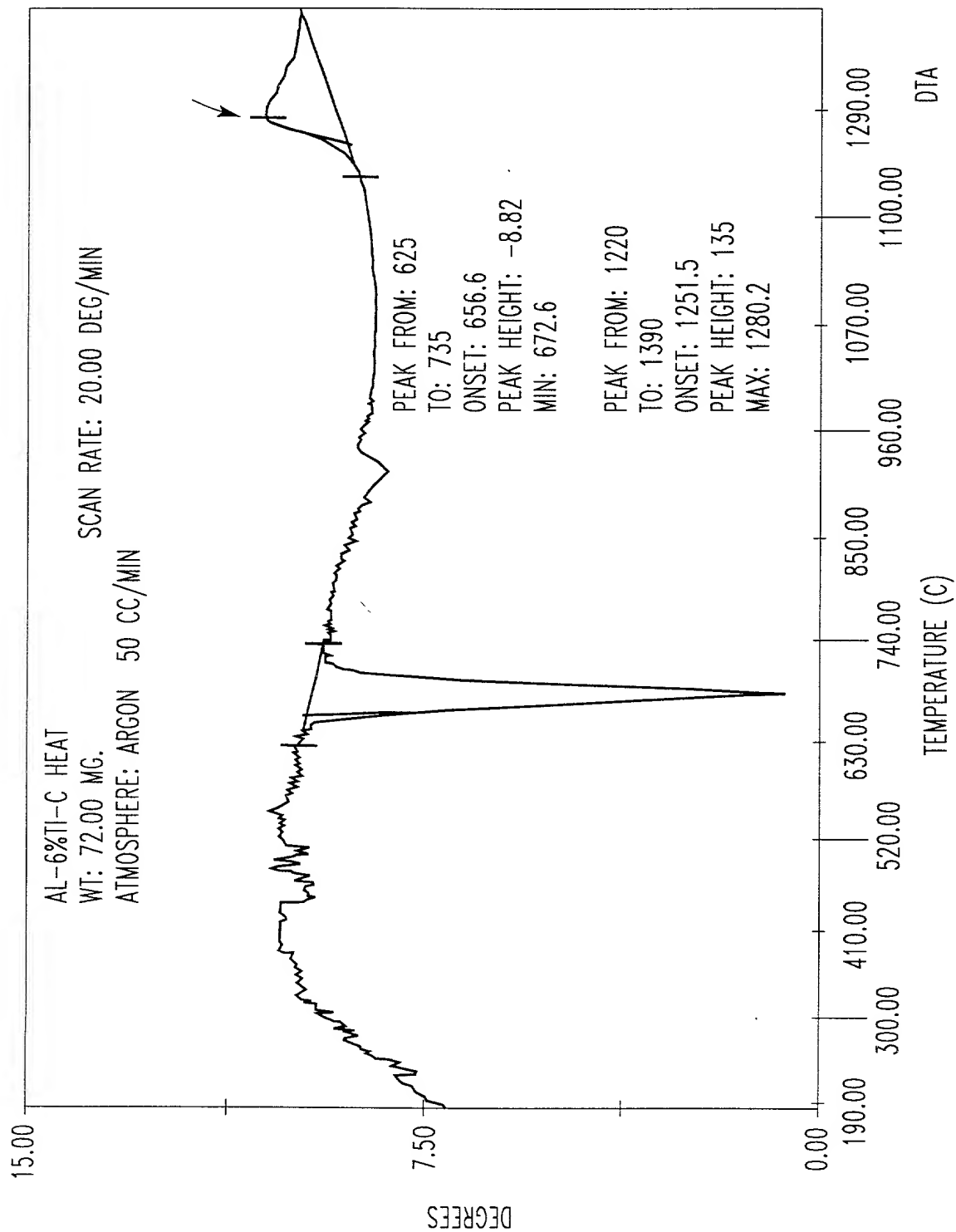
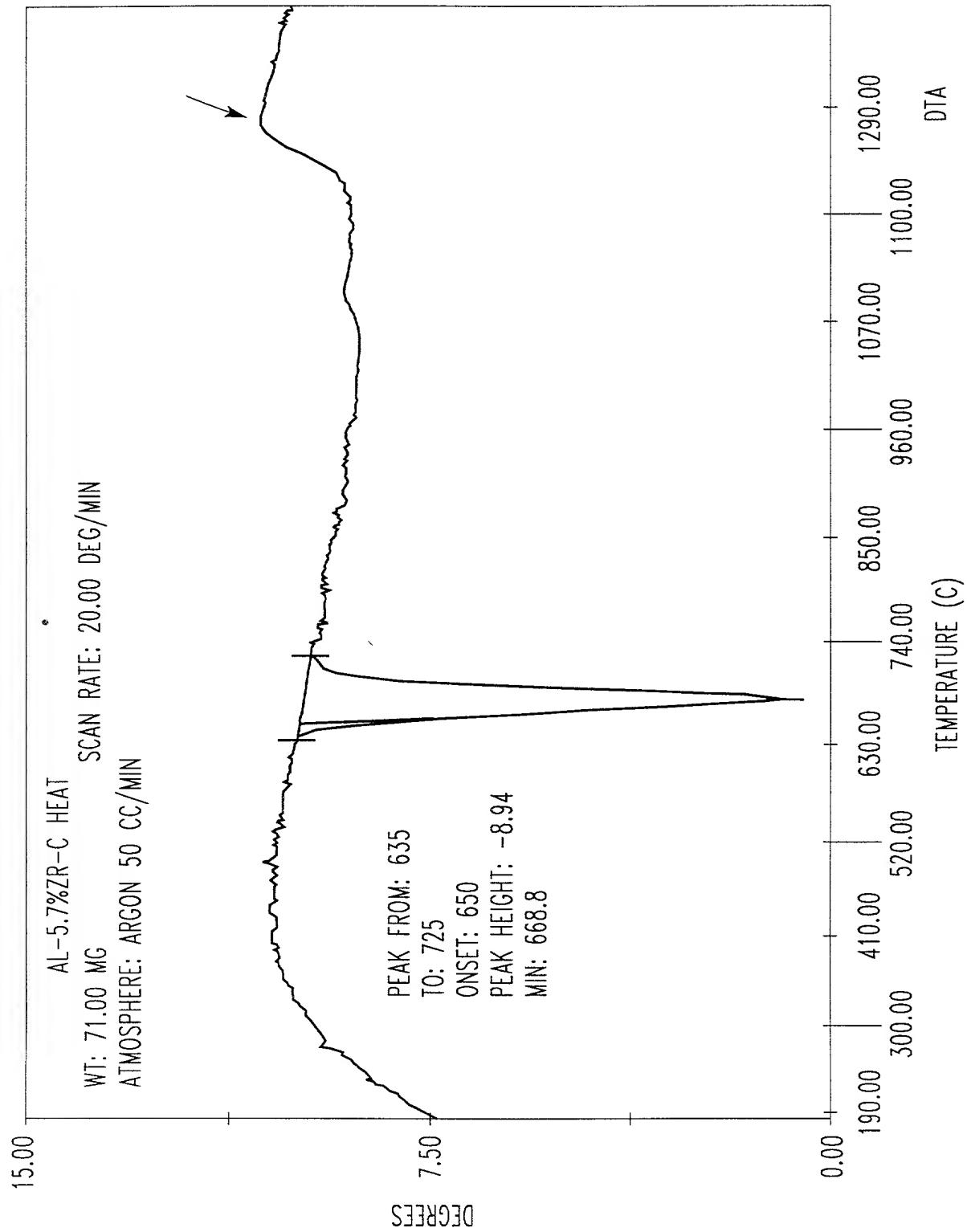


FIG.3

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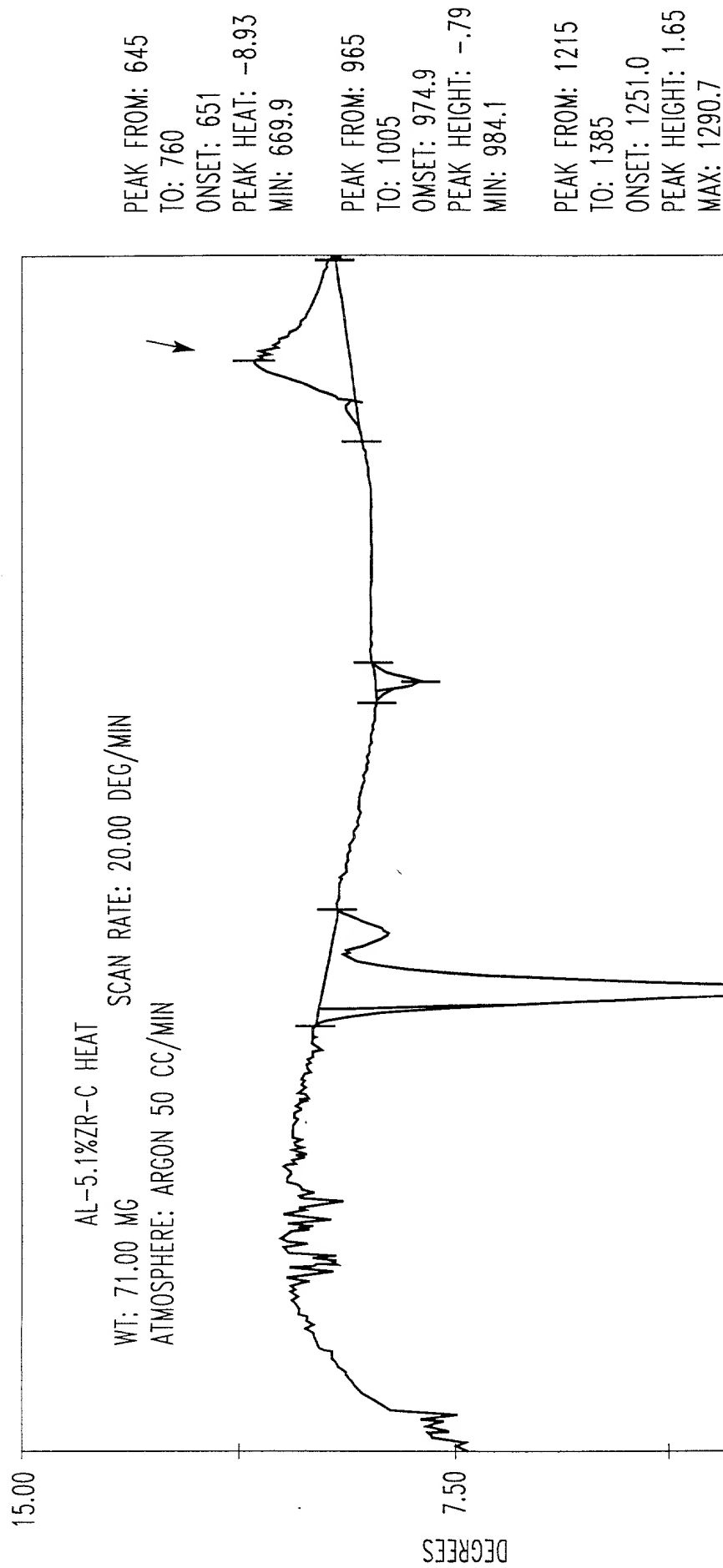


FIG.5

# DECLARATION FOR PATENT APPLICATION

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

Ceramic Particles Formed In-Situ in Metal

the specification of which ☒ is attached hereto

☐ was filed on \_\_\_\_\_ as Application Serial No. \_\_\_\_\_  
and was amended on \_\_\_\_\_ (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s)			Priority Claimed	
(Number)	(Country)	(Day/Month/Year Filed)	Yes	No
(Number)	(Country)	(Day/Month/Year Filed)	Yes	No

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application:

(Appln. Serial No.)	(Filing Date)	(Status—patented, pending, abandoned)
(Appln. Serial No.)	(Filing Date)	(Status—patented, pending, abandoned)

I hereby appoint the following attorneys to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under §1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Full name of sole or first inventor Men Glenn Chu

Inventor's signature 

Date 97-11-25

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